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Electrodialytic remediation of municipal solid waste incineration residues using different membranes

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Abstract

In the present work, three different commercial membrane brands were used in an identical electrodialytic cell setup and operating conditions, in order to reduce the leaching of metals and salt anions of two types of municipal solid waste incineration residues: air pollution control residues of a semi-dry flue-gas cleaning system and fly ashes from a plant with wet flue-gas cleaning system. The results showed a general reduction of the leaching in both residues after ED remediation. For the following elements, the leaching was found to be different after ED treatment depending on the membrane used, with statistical significance:

- Air pollution control residues of the semi-dry flue-gas cleaning system: Cr, Cu, Ni, Pb, Zn;
- Fly ashes from a plant with wet flue-gas cleaning system: Al, Ba, Cu, Ni, Zn, Cl, SO_4.

Final leaching values for some elements and membranes, but not the majority, were below than those of certified coal fly ash (e.g. Al or Cr), a material which is commonly used in construction materials; at the same time, some of these values were reduced to below the Danish law thresholds on the use of contaminated soil in constructions. These results show the potential of ED as a technology to upgrade municipal solid waste incineration residues.

Keywords: MSWI; APC residues; Fly ashes; Heavy metals; Electrodialysis

1. Introduction

Worldwide municipal solid waste generation is expected to double by 2025 [1]. Improving solid waste management is therefore an urgent priority. In many developed countries, a common practice is the incineration of solid waste, as it reduces the volume of waste by up to 90% [1] and allows energy recovery . Worldwide, approximately 120 million tonnes of waste are incinerated every year [1]. This practice generates fly ashes (FA) and air pollution control (APC) residues, originating from the treatment of the flue gas coming out of the combustion chamber, and constituting 1-5% of the original waste by mass [2]. FA and APC residues are hazardous and contain several pollutants; among them, salts and heavy metals [3].

Both residues can have different potential applications as substitution of other raw materials in production of construction materials [4]. However, such use is not allowed in some countries like Denmark, where (as in many other western countries) FA and APC residues are normally deposited after treatment [3]. At the same time, huge amounts of virgin resources are used in the construction sector. An upgrade to a higher environmental standard (like a reduced metal and salts leaching), could make them suitable for substitution of raw materials in e.g. concrete production.

Electrodialysis (ED) has been proven a promising technology for this purpose [5, 6]. ED is based on application of an electrical current to a solution or a solid suspension containing ions, forcing cations to move towards the cathode and anions towards the anode. Ion exchange membranes (IEMs), which are able to let chemical species pass depending on their charge, are strategically placed to separate cations and anions from an ion-rich compartment into different compartments. The ED setup used in this work can be seen in Fig. 1.

Previous research has focused on optimising different processing parameters for ED treatment of different materials such as contaminated soils and harbour sediments, municipal solid waste incineration (MSWI) FA/APC residues or sewage sludge ashes; important parameters being the liquid-to-solid ratio (L/S), duration of treatment, current intensity, pH as well as the number of cell compartments or the use of chemicals like H_2SO_4 [7-10]. The performance of different commercial IEMs has been compared in several ED processes like the denitrification of an aqueous solution [11], the removal of organic salts from a fermentation broth [12], as well as the production of acid from waste pickle solutions [13]. Their results show that the transport of the same ion through the different IEMs can vary substantially among the different brands: up to a threefold for Na^+ and NO_3^-, up to a 1.3-fold for organic salts, and up to a fourfold for Cl-. Two different ion exchange membrane (IEM) brands have been used in different studies of ED upgrading of MSWI APC

residues and FA: Ionics [9, 14, 15] and Neosepta [5, 6, 16]. The influence of the IEM brand used for ED treatment of MSWI residues has, however, never been investigated.

To better understand the influence of the IEM type on upgrading of APC residues and other particular materials by ED, the present work assesses the performance of four IEM trademarks on the ED treatment of MSWI residues. All IEMs were tested for treatment of two common and different waste materials (MSWI FA and APC residues) in order to evaluate if the IEMs differently affect the MSWI residue characteristics after the ED treatment. The overall aim of the investigation was to compare the leachability changes of these residues, induced by the ED treatment, among the four different IEM brands.

2. Materials and methods

2.1 MSWI residues

Two different Danish MSWI residues were studied:

I. APC residues collected from a semi-dry (**SD**) flue-gas cleaning system from the MSWI plant REFA I/S. REFA I/S incinerates approximately 120,000 tonnes of household and industrial waste annually. SD were collected on 28th of June 2011 from line 3 after APC additives (slaked lime and activated carbon).

II. **FA** collected from the electrostatic precipitator in a wet flue-gas cleaning system from the MSWI plant Vestforbrænding I/S, which is the largest plant in Denmark, incinerating approximately 520,000 tonnes of household and industrial waste annually. FA was collected in week 50 in 2011 from the ash silo 5 prior to mixing with the wet scrubber sludge.

After sampling, both residues were dried at 40°C. They were subsequently dry sieved through a 1 mm sieve, as performed in previous pilot experiments [5, 6, 16], to prevent clogging in the flow spacers when scaling up. The removed fraction represented a 1% of the total initial mass [16].

2.2 Analytical methods

Mineralogy, the metal and water soluble salt concentrations and leaching were investigated for both treated and raw MSWI residues after drying them at 105 °C. Water solubility was measured only for the untreated residues. The targeted metals were: Al, As, Ba, Ca, Cd, Cu, Cr (total Cr and Cr (VI)), Mn, Mo, Na, Ni, Pb, Zn, V, Zn; whereas the target anions were: Cl^-, SO_4^{2-} .

74 The mineralogy of each residue sample was examined using X-ray powder diffraction (XRPD) with a PANalytical
75 X'Pert PRO. Water solubility was calculated by measuring the weight loss after washing the untreated residues: 100 g
76 of each residue was shaken with 500 mL of distilled water for 5 minutes, and thereafter the supernatant was separated
77 and filtered at atmospheric pressure. This procedure was repeated two more times for the same residue before all the
78 suspension was filtered, and dried overnight at 105°C.The metal content was measured by ICP-OES (Varian 720-ES
79 ICP-OES) in triplicates after pre-treatment by Danish Standard DS259 [17]: 1 g of each residue and 20mL 7.3 M HNO_3
80 were heated at 200 kPa (120°C) for 30 min. The liquid was thereafter separated by vacuum filtration through a 45 μm
81 filter and diluted to 100 mL by de-ionised (DI) water.

82 The water soluble content of SO_4^{2-} and Cl^- was determined on IC (DIONEX DX-120 IC) in triplicates according to DS /
83 EN ISO 10304-2 [18] after water extraction. This was performed as follows: 2.5 g of each residue and 25 ml distilled
84 water were mixed, shaken for 16 hours on a horizontal shaker operating at 150 rpm, and the extractants were vacuum
85 filtered through 45 μm filters.

86 Leaching experiments were made according to DS/EN 12457-1 [19] in triplicates. The L/S was 2, mixing 40 g of each
87 residue with 80 mL DI water. The resulting suspension was shaken at 150 rpm for 24 h before vacuum filtration through
88 a 45 μm filter and the filtrate was divided into two subsamples after measuring the pH by a Radiometer Analytical pH
89 electrode. One subsample was used to measure sulphate (SO_4^{2-}) and chloride (Cl^-) concentrations on ionic
90 chromatograph (IC). The other was diluted at a ratio of 1.25 with concentrated HNO_3, and heated at 200 kPa (120°C)
91 for 30 min, before the metal content analysis on ICP-OES (Varian 720-ES ICP-OES).

92 2.3 Experimental set-up

93 The Electrodialytic (ED) cell was similar to the ones used in previous experiments [14, 20]. It consisted of four (I, II, III,
94 IV) compartments (Fig. 1) made of cylindrical Plexiglas® with an internal diameter of 8 cm. Compartment III was 10
95 cm long and contained in each experiment 35 g ash and 350 ml of DI water. An electrolyte solution (0.01 M $NaNO_3$, pH
96 < 2 adjusted with HNO_3) was circulated in each of the electrolyte compartments (I, II and IV) using Totton Pumps NDP
97 10/2. The electrolyte solution was selected based on positive results from previous researches [14, 20], where no
98 reactions were observed between the electrolyte solution and the transported species (for example, precipitate formation)
99 or the electrodes (for example, gas formation) was observed. This can also allow a more accurate calculation of the
100 mass balances explained in section 2.6. The total volumes used were 500 mL for I and IV, and 350 mL for II. A plastic

strip attached to a glass-stick connected to an overhead stirrer (IKA RW11) was used to stir the ash suspension during remediation. The electrodes were made of platinum coated titanium wire (diameter 3 mm) obtained from Permascand®. An Agilent E3612A DC power supply was used to maintain a constant electric dc current. Details on the IEMs used can be found in section 2.4.

Compartment II was included because a high concentration of soluble salts containing Cl^- was expected [3]; otherwise, these anions would enter compartment I, enabling the formation of toxic Cl_2 gas from the electrode reaction at the anode: $2Cl^- \rightarrow Cl_2+2e^-$

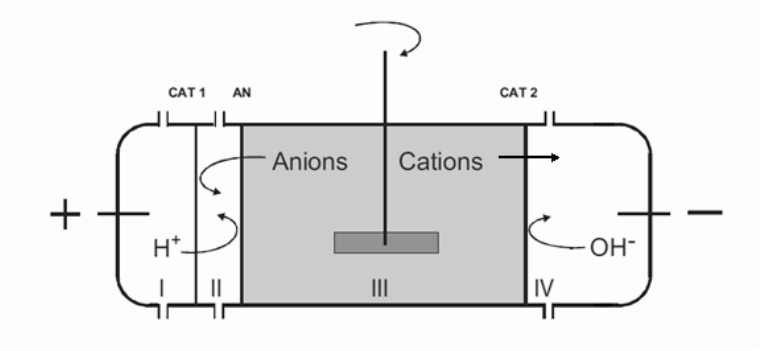

Fig. 1 Schematic view of a cell used for the ED treatment of both APC residues. AN: anion-exchange membrane; CAT1 /CAT2: cation-exchange membranes.

2.4 Membranes

Four IEM pairs were selected according to several criteria. The first two trademarks have been used previously in experimental work with ED of MSWI residues:

- Ionics (cationic: CR67-HMR, anionic: AR204-SZRA from GE), from GE Water & Process Technologies (United States), used in laboratory scale ED cell investigations to reduce the content [14, 21] or the leachability [15] of heavy metals in MSWI residues.
- Neosepta (cationic: CMX, anionic: AMX), from ASTOM Corporation (Japan), used in two pilot scale ED stack investigations to reduce the leachability of heavy metals, chloride and sulphate in MSWI residues [5, 6, 16].

The other two based on their pH range tolerance (0-14):

- Ralex® (cationic: CM-PP, anionic: AM-PP), from MEGA a.s. (Czech Republic).

- Excellion™ (cationic: I-100, anionic: I-200), from SnowPure, LLC (United States).

The pH range is a crucial criterion because of the pH differences between the compartments. MSWI residues, found in compartment III, had a considerable alkalinity [3, 15, 16]. Contrarily, the solutions in the other compartments were kept in an acidic range as explained in the section 2.5.

Common commercial applications of these IEMs are the deionisation of food products like juices or whey, water and wastewater desalination, as well as electro-coating of automotive parts. Moreover, Ionics membranes have been used in research fields like phosphorus recovery from sewage sludge ashes [10] or heavy metal removal from contaminated soils [22, 23].

2.5 Experiments

Combining the two MSWI residues studied and the four IEMs selected, eight different combinations of ED experiments were replicated until enough ED treated material was produced to carry out the analyses described in section 2.2.The duration of the ED treatments was 70 minutes at a current density of 4.67 mA/cm^2. Voltage and current between the two electrodes were monitored during the whole experiment. The pH (for all compartments) and electrical conductivity (for compartment III) were measured at the beginning, at the end, and every 20 min during the ED treatment. During the experiments, the pH of the compartment IV (Fig.1) was adjusted to <2.5 with HNO_3 1M, to avoid precipitations in the electrolyte due to the production of OH^- from electrolysis at the cathode.

Electrolyte samples were taken at the beginning and at the end of all experiments for each compartment in order to study the metal ion mobility among them. At the end of the experiments, the suspension in the central compartment was filtered at atmospheric pressure and dried during 48 hours at 50 °C. The resulting residue was loosened by hand in a mortar and stored in plastic bags under dry conditions. The volumes of the filtered liquid from the middle compartment and the electrolytes were measured. The electrodes were rinsed in 5 $mol \cdot L^{-1}$ HNO_3, whereas the IEMs and the stirrer in 1 $mol \cdot L^{-1}$ HNO_3. The samples of compartments III and IV were diluted at a ratio of 1.25 with concentrated HNO_3, and heated at 200 kPa (120°C) for 30 min, prior to the metal content analyses in all liquids on ICP-OES (Varian 720-ES ICP-OES).

147 2.6 Calculations

148 Mass balances for each studied metal, defined as the relation between the sum of mass found in the different ED cell
149 compartments at the end of the experiments and the amount initially found in the mass of untreated MSWI residue
150 placed in the ED cell, were made to control the quality of the experiments.

151 An analysis of variances (ANOVA) was run for each element and each treated MSWI residue, so as to evaluate whether
152 the leachability changes after the ED treatment was influenced by the IEM used.

153 **3. Results and discussion**

154 3.1 ED experiments performance

155 During the ED remediation with one IEM brand (Excellion), liquid flows between the compartments were observed in
156 all experiments. After contacting the manufacturer, it was determined that the membranes were degraded during the ED
157 experiments due to the use of nitrate solutions in the electrolyte compartments (I, II and IV). Specifically, it was
158 detected that a water permeability as high as 0.01327 $mL \cdot min^{-1} \cdot cm^{-2} \cdot psi^{-1}$ (the normal value would be below 0.0005
159 $mL \cdot min^{-1} \cdot cm^{-2} \cdot psi^{-1}$). Thus, the transport mechanisms of ions between compartments might not be dominated by
160 electromigration, as it can be assumed for the other three IEMs. In consequence, this IEM brand was proven not to be
161 suitable for the present ED remediation of these MSWI residues, and its results are not further analysed. The other IEMs
162 did not present major issues during the experiments.

163 The mass balances of the studied metals, defined as the recovered amount of an element in percentage of the initial
164 amount, was mainly found in a range of 80-120%. This indicates an acceptable quality of the experiments and the
165 validity of the results presented.

166 The conductivity in compartment III was stable during the ED experiments (results not shown). For the SD, the values
167 oscillated between 40 and 50 mS/cm, whereas for FA the values oscillated between 25 and 30 mS/cm, with a slight
168 tendency for decrease over the 70-minute experiments for both residues. The higher conductivity in SD than in FA
169 experiments is probably due to a higher presence of leached elements from the residues: water solubility was found to
170 be higher for SD (45.6%) than FA (12.5%). Furthermore, water soluble chloride content was almost three times higher
171 in SD than in FA before ED (Table 1). Thus, solubilised anions and cations are likely to be higher in the ED
172 experiments with SD residues. The decrease in the electrical conductivity during the experiments is due to the migration
173 of ions from the suspension into the cathode and the anode compartments. Similar trends were found in previous works

with APC residues from REFA [5, 6]. As a consequence of the electrical conductivity decrease in compartment III, voltage increased during all experiments by up to 30%. pH in compartment III was stable and between 11 and 12 during ED remediation for both residues, which is due to the high alkalinity of the MSWI residues.

3.2 MSWI residues characteristics before and after ED treatment

All XRPD diffractograms showing the mineralogy of the treated and untreated residues had a high background due to the presence of amorphous material in the MSWI residues, as previously seen in other studies [24, 25]. The identified minerals were calcite ($CaCO_3$), anhydrite ($CaSO_4$), portlandite ($Ca(OH)_2$) and Quartz (SiO_2) for untreated and treated SD. The same minerals phases, except for SiO_2, and together with Calcium chloride hydroxide (CaOHCl), Halite (NaCl) and Sylvite (KCl) were identified for untreated FA. All these phases are consistent with the results from the literature for these two kinds of MSWI residues [24-29]. After ED treatment of FA, two new main phases, (gypsum ($CaSO_4 \cdot H_2O$) and SiO_2) were found, whereas soluble phases like NaCl and KCl were not identified. Also, XRD characteristic peaks for $Ca(OH)_2$ had lower intensity for SD and were not detected for FA after the ED experiments; similarly, CaOHCl peaks disappeared after ED treatment of FA. These results indicate that a considerable part of NaCl, KCl, CaOHCl and $Ca(OH)_2$ dissolved during ED. Anhydrite intensity peak increased after all ED treatments, which can be attributed to a higher proportion due to its low solubility and the removal of other compounds.

Table 1 show the metal and the soluble salts content of both MSWI residues before and after ED. Overall, and as seen in a previous study [5], the total concentration of all metals except Na increased slightly, whereas Cl and SO_4 content were considerably reduced after the ED experiments (Table 1). These facts confirm that water-soluble salts, like NaCl, and KCl for both residues, and possibly Na_2SO_4 for FA, in the untreated residues, were dissolved and removed during ED. Despite their dissolution, Na as well as the water-soluble part of Cl and SO_4 were, together with Al, Ca, Pb and Zn, the major compounds found for both MSWI residues before and after ED treatment.

196 **Table 1** Metal and water-soluble salt anion content of SD and FA (mean value±standard deviation). The results after

197 ED are calculated as an average of the results of the three IEMs.

Compound (Units)	SD		FA	
	Before ED	After ED	Before ED	After ED
Al (g/kg)	15.5±0.09	21.4±1.11	29.4±0.36	35±0.74
As (mg/kg)	57.7±2.13	68.4±5.98	86±2.19	95.5±5.02
Ba (mg/kg)	374±6.35	409±24.2	692±19	797±49.6
Ca (g/kg)	296±3.52	301±11.7	186±2.98	198±5.84
Cd (mg/kg)	134±0.78	162±16.6	95.3±1.37	107±4.99
Cr (mg/kg)	104±0.87	125±7.54	97.2±5.17	109±4.15
Cu (mg/kg)	519±3.94	662±31.2	712±5.87	879±134
Mn (mg/kg)	352±10.4	480±13	649±10.2	739±14.2
Mo (mg/kg)	35.5±0.49	43.1±2.05	55.4±0.87	63.2±3.05
Na (g/kg)	32.5±0.68	5.38±0.37	34.6±0.32	5.88±0.11
Ni (mg/kg)	27.2±1.13	33.3±4.23	48.5±10.3	47±3.6
Pb (g/kg)	2.36±0.03	2.38±0.14	2.90±0.05	2.70±0.09
V (mg/kg)	35.1±0.78	40.7±2.61	46.2±3.01	56.7±6.04
Zn (g/kg)	15.8±0.26	21.1±1.21	20.7±0.08	23.1±1.02
Cl (g/kg)	160±2.15	15.5±1.10	60.7±1.63	2.41±0.11
SO_4 (g/kg)	14.9±0.03	10.7±0.37	34.3±0.98	14.7±0.35

198

199

200 3.3 Leaching of elements

201 3.3.1 General behaviour before and after ED treatment

202 Tables 2 and 3 show the results from the leaching tests. Category 3 values, the Danish guideline levels for water
203 leachable metals and salts for the reuse of residues as in construction materials [30], are also included. MSWI FA/APC
204 residues are generally classified as hazardous and thus these legislation limits would not be applicable, but they can be
205 used as reference to understand the upgrade made after ED experiments.

206 Ba, Pb and Zn leaching was considerably lower in FA than in SD (Tables 2 and 3), even though their content was
207 similar (Table 1). One reason can be a higher concentration of more alkaline soluble compounds containing either of
208 these elements in SD, such as $PbCl_2$, PbO [31], $ZnCl_2$ (previously identified in MSWI residues [26]) or $BaCl_2$. Another
209 explanation can be the lower pH at the end of the leaching test in FA than in SD (Tables 2 and 3): previous researches
210 showed a lower leaching of Ba, Pb and Zn at pH 11 than at pH 12 in MSWI residues [15, 32-34].

211 Overall, average leaching for metals and salts was reduced during ED treatment, a tendency also observed in previous
212 researches on ED remediation of FA and APC residues [5, 6, 15, 16]. The explanation for this trend can be the
213 dissolution and consequent removal to the electrolyte compartments (I, II and IV in Fig. 1) of the leachable part during
214 the ED treatment. Moreover, ions migration through the IEMs can enhance the dissolution of some compounds by
215 equilibrium displacement. Leaching was reduced, on average, from above to below Category 3 limits for Ba and Na in
216 SD and for As, Na, Zn, Cl, SO_4 in FA. Na and Cl leaching reduction are consistent with the decrease in total
217 concentrations (Table 1). Average leaching values were also reduced from above to below Category 3 limits for As in
218 SD regardless the IEM used, as well as for Pb in FA treated with Ralex IEMs.

219 Notable exceptions were Cr and Ni for both SD and FA, as the leaching increased after the ED experiments, in some
220 cases from below to above Category 3 limits (Table 2 and 3). The other exception was SO_4 in SD, with a leaching
221 slightly higher after ED treatment. For each element, a separate discussion is needed:

222 - The increase of Cr leaching after the experiments was also observed in previous researches for ED remediation
223 of similar FA and APC residues [5, 15, 16]. Cr-solubility in SD and FA has been suggested in other researches
224 to be controlled rather by the presence of sparingly-soluble Cr(VI) phases, like $BaCrO_4$ and $PbCrO_4$, than by
225 Cr(III) phases [26, 35]. In fact, the solubility of these Cr(VI) compounds at a high pH is higher than for Cr(III).
226 For instance, an alkaline extraction of $PbCrO_4$-spiked soils at L/S 20 released up to 50,000 µg Cr/L [23]; in

contrast, Cr(III) solubility at pH~12 is limited to around 10 µg/L[36]. The increase in Cr solubilisation after ED can be due to the removal of soluble Ba and Pb compounds, other than chromates: leaching values of Pb for both residues, and Ba in SD, were noticeably reduced after ED treatment (Tables 2 and 3). Ba and Pb ions scarcity in the leaching experiments after ED treatment can favour Cr dissolution. On the contrary, the higher concentration of Ba and/or Pb ions in the untreated residues leaching experiments may prevent the dissolution, or re-precipitate in case of release, of Ba- and Pb- chromates.

- Ni leaching increase was not generally observed before in others researches using ED to remediate similar MSWI FA and APC residues [5, 16]. The reason can be the enhanced dissolution of Al-Ni layered double hydroxide (LDH), which was found to limit Ni solubility in cement materials [37], during ED: mobilized Al during the experiments is around 20 and 50 times higher than in the leaching tests of untreated SD and FA, respectively. Thus, a higher dissolution of Al could imply a partial dissolution of Ni hydroxides, which have a higher solubility than in Al-Ni LDH.
- SO_4 leaching increase after ED treatment of SD, which was previously observed in another study for the same APC residues ([16] and Table 2), can be due to the removal of soluble Ca compounds during ED: Ca leaching in SD was considerably reduced (Table 2) after ED. The higher presence of Ca cations in the untreated than ED-treated SD leaching test can decrease $CaSO_4$ solubility in the former. After ED treatment, the lower content of Ca in the leaching tests can favour $CaSO_4$ dissolution.

3.3.2 Comparison with previous results, Category 3 guidelines and certified coal fly ash values

Compared with a previous study on the same MSWI residues [16], the leaching values before the ED treatment were within two standard deviations for all elements, except for Pb in FA which were still in the same order of magnitude (Tables 2 and 3). Leaching values for untreated SD and FA were also in the same order of magnitude for Cr, Cu, Pb and Zn as another investigation about the same residues [15]. After ED, leaching of most elements were in the range of the results in [16] (in which Neosepta was used) for SD (Table 2), and within two standard deviations for FA (Table 3). Moreover, they had the same order of magnitude for Cr, Cu, Pb and Zn than the values obtained in [15] with a 3-compartment ED cell using Ionics. Therefore, the leaching values before and after ED were in line with previous investigations.

For SD, Category 3 limits were exceeded on average for Cr, Pb, Zn and Cl after treatment regardless the IEM used and for Ni after treatment using Ionics and Neosepta IEMs (Table 2). The variability of As values was high enough for

Ionics and Neosepta IEMs, so that Category 3 limits could be exceeded or not within one standard deviation (Table 2). For FA, Category 3 limits were exceeded on average for Pb after treatment with Ionics and Neosepta IEMs; remarkably, Category 3 limits were within one standard deviation below the mean value for both IEM (Table 3). Category 3 limits were also slightly exceeded on average for Ni after ED treatment with the three IEMs. It should be noted that there was variability enough for Cr for all IEMs, for Ni with Ralex and for As with Ionics and Ralex to exceed or not Category 3 limits within one or two standard deviations (Table 3). Leaching values exceeding Category 3 for Pb, Zn and Cl were observed in a previous work with APC residues from the REFA plant [5]; in that research, the combination of the ED treatment with carbonation was successful in order to reduce the leachability for most of heavy metals except for Cr. A similar treatment could have a potential beneficial effect for the two MSWI residues of this study, although it requires further study.

Certified coal fly ash is commonly used in cement production, in contrast to MSWI APC residues, despite that its Cr leaching values normally exceeds Category 3 limits leaching values [38]. In general, considering a range of two standard deviations, ED treated SD and FA have a higher metal leaching than certified coal fly ashes. Exceptions for treated FA were Al, Ba, Ca and Cr, as their leaching in coal fly ashes is higher: 2,600 μg/L, 0.7 mg/L, 1,200 g/L and 1,000 μg/L [38], respectively (see Table 3 for comparison). For treated SD, the exceptions were Al and Cr for some IEMs (see Table 2 for comparison).

Table 2 Metal and salt leaching results from SD according to DS/EN 12457-1 (mean value±standard deviation). Average values exceeding Category 3 limits are shown **in bold**. *Below ICP limit of detection (0.02 mg/L, LOD) ** Range of average values.

	Before ED	After ED			Previous research [16]		Category 3 [28]
		Ionics	Neosepta	Ralex	Before ED	After ED **	
pH	12.3±0.04	12.1±0.01	12.1±0.01	12.1±0.08	11.9	10.2-12.2	-
As (µg/L)	**91±15**	43±16	36±4.2	48±25	<91	<20-650	50
Ba (mg/L)	**21±4.4**	1.7±0.14	1.7±0.08	1.6±0.14	**17±5**	1.6-3.0	4
Cd (µg/L)	<20*	<20*	<20*	<20*	<20	<20	40
Cr (µg/L)	64±15	**1,200±45**	**760±40**	**880±77**	61±2	114-1,015	500
Cu (µg/L)	1,200±1,500	61±4.0	55±5.7	240±15	889±397	32-141	2,000
Mn (µg/L)	100±160	<20*	<20*	<20*	<100	<20	1,000
Na (g/L)	**14±2.1**	1.1±0.04	1.2±0.05	1.1±0.09	**13±2**	**1.6-7.8**	1.5
Ni (µg/L)	<20*	**110±13**	**86±6.9**	56±5.1	<20	<20-**72**	70
Pb (mg/L)	**570±77**	**4±0.03**	**3.5±0.81**	**14±1.3**	**453±168**	**1-21**	0.1
Zn (mg/L)	**39±3.9**	**4.5±0.13**	**4.4±0.45**	**6±0.41**	**32±10**	0.1-**3.7**	1.5
Al (µg/L)	930±120	360±71	270±14	220±280	-	-	-
Ca (g/L)	32±4.7	4.8±0.13	4.9±0.2	4.7±0.43	-	-	-
Mo (µg/L)	770±57	600±23	600±16	550±50	770	211-708	-
V (µg/L)	<20*	<20*	<20*	<20*	<20	311-662	-
Cl (g/L)	**85±1.7**	**8.3±0.25**	**8.9±0.26**	**8.7±0.36**	**87±3**	**5.1-35**	3
SO_4 (g/L)	0.87±0.03	1.3±0.03	1.3±0.01	1.3±0.03	0.9±0	0.9-1.2	4

Table 3 Metal and salt leaching results from FA according to DS/EN 12457-1 (mean value±standard deviation). Average values exceeding Category 3 limits are shown **in bold**. *Below ICP LOD. ** One of the triplicate measurements value below LOD, so the result is shown as an average of the other two triplicates and LOD/2.

	Before treatment	After treatment			Previous research [16]			Category 3 [28]
		Ionics	Neosepta	Ralex	Before ED	After ED		
pH	11.3±0.24	11.1±0.01	11.1±0.01	11.0±0.03	12.6	10.1±0.1	11.9±0.4	-
As (µg/L)	**59±20**	40±14	36±2.8	48±22	<20	<20	<29	50
Ba (mg/L)	0.46±0.05	0.29±0.02	0.3±0.003	0.22±0.02	0.4±0.03	0.8±0.9	2.6±0.1	4
Cd (µg/L)	<20*	<20*	<20*	<20*	<20	<20	<20	40
Cr (µg/L)	18±7.5**	430±44	480±49	460±46	20±0	**991±234**	341±146	500
Cu (µg/L)	30±6.4	26±1.7	20±3	16±3.5	23±11	37±14	28±25	2,000
Mn (µg/L)	<20*	<20*	<20*	<20*	<20	<20	<20	1,000
Na (g/L)	**12±0.8**	0.43±0.02	0.46±0.02	0.5±0.04	**14±2**	**3.6±1.1**	**3.1±0.6**	1.5
Ni (µg/L)	<20*	**130±21**	**140±7.7**	**75±8.4**	<20	<**193**	<47	70
Pb (mg/L)	**18±2.4**	**0.47±0.74**	**0.13±0.13**	0.03±0.01	**40±2**	**1.8±2.2**	**18±11**	0.1
Zn (mg/L)	**4.5±0.67**	0.57±0.26	0.46±0.13	0.27±0.02	**3.3±1.1**	0.2±0.0	**3.5±2.8**	1.5
Al (µg/L)	610±44	1,100±130	1,000±96	150±14	-	-	-	-
Ca (g/L)	1.4±0.07	0.97±0.01	0.98±0.01	0.93±0.06	-	-	-	-
Mo (µg/L)	2,000±150	640±27	660±14	630±48	2,024±48	704±76	497±16	-
V (µg/L)	<20*	<20*	<20*	<20*	4±55	**135±177**	**264±339**	-
Cl (g/L)	**31±0.21**	1.4±0.03	1.4±0.02	1.6±0.06	31±1	**14±11**	**20±7**	3
SO_4 (g/L)	**8.4±0.30**	1.6±0.004	1.7±0.04	1.8±0.04	9±0.5	1.9±0.4	1.0±0.4	4

278 3.3.3 Difference in leaching values among the IEMs

279 The result of the ANOVA among the different ED treatments for each residue with a P-value>0.05 showed that mean
280 leaching values were found not to be the same for some of the elements among the three IEMs studied. More
281 specifically, mean leaching values of Cr, Cu, Ni, Pb and Zn for the treated SD, and Ba, Cu, Ni, Al, Cl and SO_4 for the
282 treated FA were found to be different depending on the IEM used. It is worth mentioning that Pb and Zn leaching
283 values had a high variability for FA treated with Ionics and Neosepta IEMs (Table 3), which requires further study. The
284 reason could not be the pH of the different leachates, since they were almost identical regardless the membrane used
285 (Tables 2 and 3).

286 Main differences were found for Pb in SD: leaching after ED treatment with Ralex was around 3 to 4 times higher than
287 with the other two IEMs (Table 2). The reason can be a different transportation rate of chloride anions through the IEMs.
288 Ralex IEMs are likely to transport Cl^- at a slower rate than Ionics and Neosepta. This can be seen in the water soluble Cl
289 content for both residues, which was slightly higher after ED treatment with Ralex than with Ionics or Neosepta: 17.0,
290 15.6 and 15.0 mg/kg for SD and 2.54, 2.36 and 2.32 mg/kg for FA, respectively. A slower migration of Cl through
291 Ralex IEMs can entail a lower dissolution of $PbCl_2$ by equilibrium displacement, and, consequently, the content of
292 sparingly soluble $PbCl_2$ in SD after ED treatment with Ralex can be higher than when using Ionics or Neosepta.
293 However, the transportation of Cl^- and the other species through the different IEM brands, and its connection to
294 differences in leachability, require specific research for proper confirmation.

295 **4. Conclusions**

296 The leaching of some heavy metals and salts in two MSWI residues was found to be different after ED treatment
297 depending on the IEM brand used, without achieving a better overall result for any of them. Further investigation is
298 required in order to understand the unalike transportation of chloride and other ions among different IEMs during ED,
299 and its influence in the leaching behaviour of the hazardous elements from MSWI residues.

300 ED remediation resulted in the reduction of the leachability for most heavy metals and salts, probably due to the
301 removal of the leachable part. For some metals, like Al or Cr, the leaching was reduced to below the values found in
302 certified coal fly ash. At the same time, leaching was reduced for some elements like As from above to below the
303 Danish guideline levels for the use of residues as construction materials. However, none of the resulting ED-treated

MSWI residues had all the leaching values below these levels, and for most elements the final values were higher than for certified coal fly ash.

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